Critical Experiment with Uranium Diluted with Concrete and Polyethylene

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ABSTRACT

An experiment has been performed combining highly enriched uranium, a hydrogenous moderator (polyethylene), and concrete. The purpose of the experiment was to provide additional criticality data that can be used to verify and validate criticality safety evaluations in support of the decommissioning of nuclear facilities throughout the Department of Energy complex. In this experiment, criticality was observed as a function time due to the curing and drying processes that occurred in the concrete.

INTRODUCTION

A critical experiment containing concrete, polyethylene, and highly enriched uranium (HEU) was performed as part of the decommissioning of nuclear facilities program. To the best of our knowledge, few critical experiments that contain concrete as a moderator have been performed to assure the correctness of the calculations used for criticality safety evaluations. The experiment was performed on three different occasions using concrete that had been poured 1 day before, 6 months before, and 7 months before to reveal the effects that curing might have on the criticality of the experiment.

The experiment was designed to study how the critical mass changes, as a function of time, due mainly to the curing and drying processes of the concrete. The experiment was designed to be in the undermoderated region so that if water from the concrete were to evaporate, the resulting configuration would be less reactive. This means that more uranium would be needed to attain criticality as water evaporates from the configuration. The results presented in this paper establish the critical masses as a function of the curing process of the concrete. This experiment will help validate cross-section data used in computer neutron transport codes.

DESCRIPTION OF THE EXPERIMENT

The experiment was fueled with HEU foils and performed on the Planet vertical assembly machine [1]. The average dimension of the bare foils was 22.86 cm square by 0.00762 cm thick. The foils were laminated with plastic sheets. Each foil weighed approximately 70 g, and their isotopic compositions were 93.23 wt % ^{235}U, 5.37 wt % ^{238}U, 0.26 wt % ^{236}U, and 1.13 wt % ^{234}U. The moderating polyethylene plates were 39.12 cm square by 1.91 cm thick. Each moderating polyethylene plate had a central recess that was 22.86 cm square by 0.64 cm deep.

Before the concrete mix was prepared, pebbles larger than 0.64 cm in diameter were screened out. The concrete mixture was prepared by adding 0.14 L of water to each kilogram of concrete mix. The concrete mixture was then poured into the recess of each moderating polyethylene plate (Fig. 1). The density of the concrete was calculated by weighing the moderated polyethylene plate before the concrete was added and again after the poured concrete had solidified.

The plates were weighed on three different occasions. As shown in Table I, the weight of the plates changed as a function of time because of the curing process of the concrete, and in some cases, the difference in weight was as much as 10 g. The concrete density was obtained by dividing the difference in the weight of the polyethylene plate by the recess volume. The average density of the concrete poured in the recess area of each moderating plate was 1.80 g/cm^3, based on the last weight measurement of the plates performed on October 2003. This density was a bit lower than the theoretical density for concrete, which could indicate that the recess volume in the polyethylene plate was not completely filled with concrete and/or that the concrete mix excluded pebbles larger than 0.64 cm in diameter. Figure 2 shows the moderating plates with concrete inserts one day after they were poured.

Figure 1. Concrete being poured into the recess of the polyethylene plates.

Table I. Weights of Some of the Moderating Plates Containing Concrete

	March 03	**September 03**	**October 03**
Plate #48	4093.9 g	4082.7 g	4082.8 g
Plate #46	3087.7 g	3076.9 g	3076.3 g
Plate #33	3068.8 g	3059.4 g	3059.1 g
Plate #30	3089.6 g	3080.5 g	3079.9 g
Plate #22	3052.4 g	3045.8 g	3045.5 g

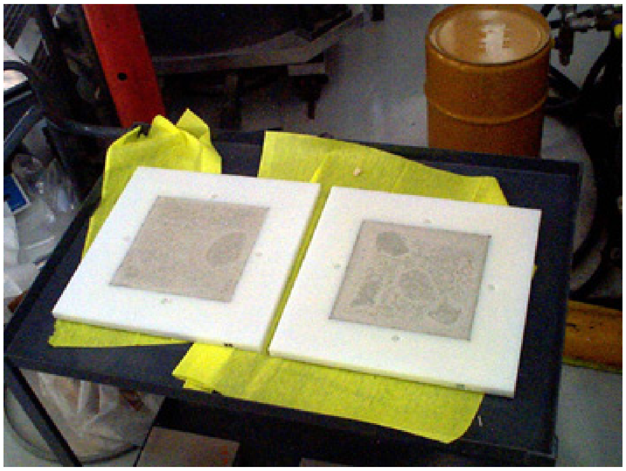

Figure 2. Moderating polyethylene plates with concrete inserts one day after pouring.

Three concrete samples were taken from each of the concrete inserts in three moderating polyethylene plates. These polyethylene plates were located at the top, in the middle, and at the bottom of the core. The chemical compositions of the concrete samples, shown in Tables IIa and IIb, represent their compositions as of October 2003. The results shown in these tables are averages of the nine concrete samples that were analyzed. The results have an uncertainty of ±5%. It is important to note that impurities of boron and cadmium were found in the concrete samples that were analyzed. These impurities are important because cadmium and boron are both thermal neutron absorbers that will have a negative reactivity effect in these experiments.

The unit cell for this experiment was defined as 22.86 cm by 22.86 cm. Each unit cell contained two uranium foils. The HEU foils were placed on each moderating polyethylene plate containing the concrete matrix material. There were eight 39.12 cm square by 2.54 cm thick high-density polyethylene plates that formed the top and bottom reflector (four at the top and four at the bottom).

Table IIa. Composition of Concrete

Chemical Composition	**Typical Content (wt %)**
Water (H_2O)	1.0744
Calcium Oxide (CaO)	14.095
Silica (SiO_2)	66.49
Iron Oxide (Fe_2O_3)	1.95
Aluminum Oxide (Al_2O_3)	8.36
MgO	0.81
Sulfate (SO_3)	0.71
Na_2O	1.40
K_2O	2.97
TiO_2	0.28
P_2O_5	0.10
SrO	0.08

Table IIb. Measured Impurity Content of Concrete

Chemical Composition	**Amount (μg/g)**	**Chemical Composition**	**Amount (μg/g)**
Ag	10	Nd	24
As	50	Ni	20
Au	10	Os	10
B	54	Pb	22
Ba	797	Pr	10
Bi	10	Pt	500
Cd	82	Rb	90
Ce	62	Re	100
Cr	112	Rh	433
Co	10	Ru	10
Cs	10	Sb	18
Cu	27	Sc	500
Dy	10	Se	100
Er	10	Sm	10
Eu	10	Sn	18
Ga	41	Ta	500
Gd	10	Tb	10
Ge	50	Te	10
Hf	50	Th	12
Hg	50	Tl	10
Ho	10	Tm	10
In	50	U	10
Ir	10	V	106
La	29	W	500
Li	160	Y	29
Lu	10	Yb	9
Mn	390	Zn	37
Mo	35	Zr	190
Nb	500		

RESULTS

1/M curves were plotted based upon the normalized counting rates as a function of the number of unit cells and as a function of separation distance. The $H/^{235}U$ ratio for this experiment was 158 and considers only the polyethylene in the experiment. As mentioned above, chemical analysis for the concrete was performed using the concrete from the October 2003 experiment. Figure 3 illustrates the setup of the critical experiment on the Planet assembly. The experiment was performed three times (March, September, and October 2006) using the same plates and HEU foils. Approximately half of the critical mass is on the movable platen and the other half is on the top platform. Monte Carlo N-particle (MCNP) [2] models were developed to benchmark the critical mass experiment, which was performed on October 28, 2003 [3]. The total amount of concrete in this experiment was 10914 g. The amount of polyethylene in the core section of the experiment was 10842 g. Table III lists the parameters of the measured critical configurations.

To determine the uncertainty in the experimental k_{eff}, MCNP calculations were performed by varying the mass of the concrete. This was accomplished by changing the density of the concrete by the relative uncertainty of the total mass of concrete (1.37%). The resulting uncertainty in k_{eff} was ±0.00065. The other calculation involved estimating the uncertainty in k_{eff} due to the dimensional uncertainty in the concrete. This was accomplished by varying the dimensions of the recess in the polyethylene on the x- and z-planes. For the thickness, the dimension was varied by 1%, which is the best estimate of the thickness uncertainty. The dimensions were varied one dimension at a time. The effect of the dimensional uncertainty in each direction was combined quadratically to get an estimate of the standard deviation. The resulting standard uncertainty in k_{eff} due to the uncertainty in the concrete dimensions was 0.0004. Based on these calculations, the uncertainty in the experimental k_{eff}, due to the uncertainty in the concrete mass and its dimensions for the experiment performed in October 2003, was 0.00076. Although no calculations were run for the same experiment performed in March and September 2006, it is estimated that the uncertainty in the experimental k_{eff} was the same as for the experiment performed on October 2003.

Figure 3. HEU-concrete experiment mounted on the Planet assembly.

The critical masses reported in Table III were obtained at one day, six months, and seven months after the concrete was poured. The same polyethylene plates and HEU foils were used for these experiments. The position of the plates and HEU foils was also the same for the three experiments. As Table III illustrates, the experiment performed on March 13, one day after pouring the concrete into the moderating polyethylene plates, contains less HEU uranium and is more reactive than the experiments performed later. The experiments performed at six and seven months contain the same amount of HEU and their experimental k_{eff} are the essentially the same, if we consider the standard uncertainty in the k_{eff}. Both of these experiments were less reactive than the first experiment. Thus, it appears that the criticality for these experiments is time dependent due mainly to the curing and drying process that occurs in concrete as a function of time. Because this experiment was designed to be undermoderated, the loss of water due to the drying process caused an increase in the critical mass, as seen in Table III. It is important to note that the chemical reaction of water with cement in concrete is extremely important to the properties of concrete and may continue for many years. Therefore, we intend to design critical experiments where the concrete will be the only moderating material to observe if the critical mass of these experiments also changes as a function of time.

Table III. Critical Parameters of Uranium Diluted with Concrete and Polyethylene

Date Experiment Performed	**$H/^{235}U$** (Considers only the hydrogen in the polyethylene)	**Mass of Uranium (g)**	**Experimental k_{eff}**
March 13, 2003	$H/^{235}U = 158$	2537.0 ± 2.25	1.0023 ± 0.00076
September 18, 2003	$H/^{235}U = 158$	2604.0 ± 2.27	1.0004 ± 0.00076
October 28, 2003	$H/^{235}U = 158$	2604.0 ± 2.26	1.0001 ± 0.00076

REFERENCES

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